

This Page Is Inserted by IFW Operations  
and is not a part of the Official Record

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images,  
please do not report the images to the  
Image Problem Mailbox.**

103 type

224322

COMMONWEALTH OF AUSTRALIA  
**PATENT SPECIFICATION**

36, 398/58

Complete Specification Lodged ..... 26th March, 1958.

Application Lodged (No. 36, 398/58) ..... 26th March, 1958.

Applicant ..... Fabriques de Produits Chimiques de Thann et des  
Mulhouse.

Actual Inventor ..... Raymond Holbein.

Convention Application.  
(France, 28th March, 1957).

Complete Specification Published ..... 25th September, 1958.

Complete Specification Accepted ..... 1st October, 1959.

Classification 79.2.

No drawing.

**COMPLETE SPECIFICATION.****"ORGANOPHILIC AND HYDROPHOBIC PIGMENTS AND METHOD  
FOR THEIR PREPARATION."**

The following statement is a full description of this invention, including the best method of performing it known to us.

The present invention relates to pigments easily wetted by organic compounds such as solvents, paint vehicles and plastic material of various nature; and to methods for their preparation.

It is generally difficult to incorporate most pigments in paint compositions. Prolonged and expensive grinding is therefore necessary to obtain a thorough dispersion of these pigments. Even so such dispersions are often of poor stability and on storage the pigment may settle as a deposit which is not easily re-suspended. Paints prepared with such pigments generally lack fluidity, a fact which limits the amount of pigment that can be incorporated without imparting too high a viscosity to the paint compositions. Also paint films containing such pigments usually lack regularity, gloss and smoothness. For the same pigment these drawbacks vary with the nature of the associated vehicle. They are especially noticeable when mineral pigments such as lithopone, zinc oxide or titanium

---

224.322

---

dioxide, either rutile or anatase, are used with vehicles containing large amounts of synthetic resins. All these drawbacks are essentially due to the lack of physical compatibility between these pigments and the non polar organic substances associated therewith.

The mineral pigments such as lithopone, zinc oxide, titanium dioxide usually present a hydrophilic surface, that is, a surface easily wetted by water in which they can be readily dispersed. This property, the "hydrophilic" nature, usually is on a par with a difficult wetting by organic liquids, such as carbon tetrachloride, benzene, fatty or mineral oils and the like; therefore it is difficult or even impossible to obtain stable dispersions of hydrophilic pigments in such organic solvents. These pigments may be called "organophobic". Other pigments by their nature are easily wetted by organic media and do not give rise to the troubles encountered with the organophobic pigments or at least to a much lower degree. They may be called "organophilic". The latter are usually wetted by water with difficulty and thus are named "hydrophobic".

Together with the property of easy preparation, paints containing organophilic pigments show other advantages, so the oil absorption of the pigment is noticeably reduced. Due to better wetting of the pigment by the vehicle, the paint film is tighter, less porous and its protecting effect is increased. An association of an organophilic and hydrophobic pigment with a suitable vehicle will produce wholly hydrophobic paint films which prove highly resistant to water and outside exposure. Such films are especially resistant to degradation by sea water when compared to films containing hydrophilic pigments.

Emulsified paints consist of a stable dispersion of a pigment and a resinous binder in an aqueous medium. After the evaporation of the water the paint film is formed by agglutination of the remaining components. Such a film will be tight and glossy only if the pigment is perfectly compatible with the resinous binder. Indeed the resin must fill all the interstices between the pigment particles and consequently must come in close contact with most of the pigment surface which requires thorough wetting abilities between the pigment and the resin, otherwise the film will be of a loose texture, show a dull surface and offer a poor protection.

Various pigments are incorporated in a great number of plastic materials for colouring or other purposes. For the reasons explained above organophilic pigments are particularly suited for incorporation in such plastic material.

Organophilic and hydrophobic pigments also find wide uses in the preparation of printing inks. These inks consist of a liquid of organic nature in which a pigment is dispersed. They must be very opaque and therefore require a high pigment to vehicle ratio and nevertheless must

---

224.322

---

remain sufficiently fluid for the inking process. In this connection organophilic pigments present great advantages. Furthermore an hydrophobic pigment is highly desirable especially in lithography where only inks of strongly hydrophobic properties can be used successfully.

In the production of synthetic fibers, such are obtained by extrusion of organic polymer substances either molten or dissolved in a suitable solvent. These fibers are usually delustered by incorporating a white pigment in the plastic mass to be extruded. In this case organophilic pigments offer a very great advantage owing to an excellent dispersion which is a determining condition for the tensile strength and the uniformity of the fibers.

Other uses for organophilic and hydrophobic pigments will be found in the preparation of cosmetic and pharmaceutical compositions of a greasy nature and also in numerous cleaning and preservative products such as for instance the pastes for white leathers and fabrics which are made of a suspension of white pigments in organic vehicles.

Numerous processes have been disclosed for imparting organophilic and hydrophobic properties to various substances and more specifically pigments which normally lack these properties or exhibit them only in a minor degree. These processes usually apply to mineral pigments but may also be used for the treatment of certain coloured organic pigments, which owing to the presence of highly polar functions in their molecule, are only weakly organophilic. Among the mentioned substances are for instance titanium dioxide, either anatase or rutile, metal titanates, zirconium oxide, zircon, blanc fixe, magnesium silicate, clays, lithophone, barium carbonate, silica, aluminium silicates, zinc sulfide, zinc oxide, antimony oxide, white lead, aluminium oxide, magnesium fluoride, carbon black, ultramarine blue, chrome yellow, basic zinc chromate, chrome red, chrome orange, barium chromate, chrome green, prussian blue, lacques of acid dyes precipitated on aluminium oxide and similar products.

All these known processes aim to coat the surface of the pigments with a thin layer of an "active substance" which imparts the organophilic property. This coating is sometimes referred to as a monomolecular film. The "active substances" used are characterized by non polar hydrophobic molecules with strongly polar substituents attached thereon; the basic non polar molecules are always hydrocarbon rests and generally but not necessarily of aliphatic nature. It is generally assumed that these molecules are deposited on the surface of the pigment with their polar substituents in close contact to that surface. Under these conditions the non polar hydrocarbon radicals are rejected away from the pigment to "active substance" interface. Thus the outer surface of the particle pigment plus "active substance" consists of hydrocarbon radicals and for

224, 322

this reason the particle acts in dispersion media as if it were entirely of hydrocarbon nature: the particle is typically organophilic and hydrophobic.

Various substances have been described for use as "active substance" such as: fatty acids with more than 8 carbon atoms and their salts; resinic acids and their salts; naphthenic acids and their salts; waxes; sulphonated vegetable or animal oils; lipoids like cholesterol, lecithine or the like; isocyanates with a long carbon chain; long chain amines and their derivatives; long chain quaternary ammonium salts and long carbon chain products of the following types: betains, sulphonium- and phosphonium salts; phthalic acid and its derivatives; substituted thioureas, metallic dialcylodithiocarbamates; rubber vulcanisation accelerators, substituted polysiloxanes; polymerisable alcoylmethylolmelamines. "Active substances" of another type include compounds containing an hydrocarbon radical associated with a substituant capable of reacting with removable hydrogen atoms especially those of hydroxyl groups which may exist on the pigment surface or may have been created thereon by suitable means. Among these substances are: acid anhydrides; acylchlorides, aliphatic and aromatic isocyanates and organo-substituted chlorosilanes.

In fact while it is relatively easy to select a suitable "active substance" it is difficult to obtain an even coating thereof on the surface of the pigment particles.

Processes have been suggested by which the pigment is dispersed in an organic solvent containing the "active substance" in solution. After this treatment the solvent is removed by evaporation and the pigment is disintegrated by mechanical means. These prior art processes are expensive due to the unavoidable solvent losses, furthermore the treatment of large amounts of pigments by such processes involve serious health and fire hazards. In the case of titanium dioxide pigment there often develops a yellow discolouration during the solvent evaporation which, as in the case of toluene requires temperatures as high as 120°C for complete elimination. Moreover these types of processes meet with an unavoidable inherent difficulty. The pigment to be treated is organophobic and therefore repellent to the solvent in which it is not adequately dispersed. Numerous pigment particles remain in an agglomerated state and consequently are not fully coated by the "active substance"; this results in an incomplete treatment, that is, an heterogeneous product. It has therefore been suggested to perform this type of treatment in grinding mills in the presence of organic solvents, which is of course an expensive operation.

Other prior art processes imply the treatment by the "active substance" in an aqueous medium. The "active substance" is added as an emulsion to a slurry of the pigment in water. Under these

224,322

conditions the "active substance" associates with the pigment due to its affinity for the pigment to which it confers the desired properties. The water is then removed and the pigment is disintegrated. Such a method gives irregular results and it has been recognized that special precautions must be taken to ensure uniform coating of all the particles. An improvement has been suggested which consists in adding a dispersing agent to the pigment slurry before the treatment with the active substance. In other words the pigment is first made strongly hydrophilic to secure its dispersion in water. The so dispersed pigment will offer the whole surface of its particles to the "active substance" treatment. This results in a uniform and reproducible coating treatment by the "active substance".

In the specific case of titanium dioxide pigments it has been suggested to add the "active substance" to a pigment which has been previously "surface treated". Surface treated pigments means pigments treated by processes in which the individual particles of the pigment are coated with a thin film of a colourless mineral compound. For instance hydrous oxides or some other compounds of Al, Mg, Zn, Ti, Zr, Ce and the like, have been suggested. These surface treatments improve the pigmentary properties of the so coated titanium dioxide.

According to a known process a titanium dioxide pigment is surface treated as referred to above, then washed, filtered and dried. Afterwards it is rendered organophilic by treatment with an "active substance" in an organic solvent.

✓ It is an object of the present invention to provide a process for treating mineral pigments and more particularly titanium dioxide pigments in a novel and simple manner by both "active substances" and mineral treating agents.

It is another object of the present invention to produce highly organophilic and hydrophobic pigments.

Another object of the invention is the production of titanium dioxide with highly improved pigmentary properties.

Still another object of the invention is to perform the treatment by the "active substance" and by the mineral treating agent without intermediate washing and drying.

In using the process hereinafter described the advantages previously separately obtained by two distinct treatments are fully secured by a single treatment. The "active substance" is hereby particularly well fixed on the pigment. In some cases and quite unexpectedly some advantages due to the mineral coating are enhanced when compared to the results obtained by known mineral coating processes. This was made possible by applying to the pigment first the treatment by the "active substance" and only subsequently the treatment by metallic salts. *Subst*

---

224,322

---

The present invention relates to the preparation of pigment of high organophilic and hydrophobic properties. More specifically it relates to the preparation of titanium dioxide containing pigments of organophilic and hydrophobic properties further characterized by high tinting strength, excellent brightness and showing exceptional resistance to chalking, to yellowing by light and to baking discolouration when incorporated in paint compositions.

In brief, the method according to the invention consists in dispersing the pigment in water in the presence of a dispersing agent, adding to this suspension "the active substance" conferring organophilic properties and adding to the mixed dispersion obtained a solution of a metal salt capable of giving a mineral coating and simultaneously suppressing the action of the dispersing agent. Subsequently the product is as usual, neutralized, filtered, washed, dried and disintegrated.

The conventional titanium dioxide production processes generally include a hydroclassification step after the grinding of the calcined product. According to the present invention, a suspension of such hydroclassified pigment will preferably but not necessarily be used as the material to be treated. In certain cases an adjustment of the concentration of the pigment and/or of the dispersing agent in that hydroclassified suspension may be useful.

Some results may be obtained using pigment suspensions free of dispersing agents, but it has been observed that the results are so much the better and the more regular as the dispersion has been more perfect, for that reason the use of dispersing agents is highly useful. The amount of dispersing agent to be employed depends primarily on the chemical nature of this agent but must also be varied according to the nature of the pigment to be treated. Usually the required amount will be between 0.03% to 3% by weight of the dry pigment.

According to the present invention, the selection of the appropriate dispersing agent is directed by the necessity of compatibility with the "active substance", it being required that the dispersion remains stable after additions of the "active substance". Furthermore the dispersing agent should not disturb the action of the metal salt to be added in the third step; on the contrary it is desirable that the dispersing agent contributes with the metal salt to produce additional improvements of the pigment properties. In the case of titanium dioxide, soluble silicates, especially sodium and potassium silicates, are very suitable as dispersing agents. Good results have also been obtained by using alkali metal phosphates and polyphosphates or mixtures of arabian gum and saccharose.

"Active substances" conferring to the pigments the desired organophilic properties are numerous. When selecting the appropriate

---

221,322

---

"active substance" the following limitations will be taken in account: it should be compatible with the dispersing agent used; it should adhere tightly to the considered pigment and moreover it should show good resistance to the destructive agents to which the pigment is likely to be exposed such as outside conditions of heat, light and chemicals, for example. It is advantageous to add the "active substance" in form of a fine dispersion in an aqueous medium: either as a true or colloidal solution, or as a solution in a water miscible solvent or as an aqueous emulsion of the "active substance" either pure or dissolved in a water immiscible solvent.

All the products specified above as "active substances" may be used according to the present invention. Two classes of substances have been found particularly suitable:

- a) the alkali metal salts of the higher fatty acids, of resinic acids and of naphthenic acids all used as aqueous solutions or as aqueous sols.
- b) mono and disubstituted polysiloxanes, the substituents being alkoxy- aryl or aralkoxy radicals either saturated or ethylenic.

These water insoluble substances are used in the form of stable aqueous emulsions.

While noticeable results may in some cases be obtained by using as little as 0.1% of "active substance" the usually employed amount lies between 0.5% to 2% based on the dry weight of the pigment; it is however, possible in special cases to fix up to 5% "active substance" on the pigment. Obviously the necessary quantity of "active substance" depends largely upon the surface area of the pigment, that is, essentially on the mean particle diameter, the figures given above being those for particles in the one micron range. If desired a mixture of several "active substances" may be used; in this case the total weight of these substances amounts to the above given figures.

It is important that the dispersion containing the pigment and the "active substance" is a stable one in order to secure a uniform action of the metal salt added in the next step. This is the case if dispersing agent and "active substance" are compatible. As previously mentioned, water insoluble "active substances" are advantageously used in form of aqueous emulsions. In these cases care must be taken to avoid that the emulsion is made up with emulsators which would be incompatible with the dispersing agent of the first step. Otherwise it would be difficult if not impossible to secure an even distribution of the "active substance" on the pigment and still more difficult to ensure a proper action of the metal salt added in the next step. Anionic, cationic or non ionic emulsators may be used and experiment will permit selection of the most suitable, the criterion being the stability of the pigment plus "active substance" mixed dispersion.



---

224.322

---

In order to secure a perfect distribution thereof, the "active substance" will preferably be added progressively to the pigment suspension under good stirring, and mixing will be continued as long as necessary.

In the next step addition is made to the mixed dispersion obtained as described, of a metal salt able to deposit on the surface of the pigment a layer of a colourless compound. Salts of the following metals may be used: aluminium, magnesium, zinc, titanium, zirconium, the metals of the rare earths group, tin, antimony, lead, as well as mixtures of salts of one or several of these metals. In practice soluble salts or aluminium will as an example secure excellent results. The addition of the metal salt, advantageously as an aqueous solution, will preferably be done progressively and under good stirring, which stirring will be continued for some time. The action of the added metal salt is of various nature. First by its hydrolytic decomposition, the salt liberates acidity which lowers the pH of the suspension thus decreasing the action of the alkaline dispersing agents. The hydrolysis results in hydrous metal oxide which is deposited on the pigment surface in common with the "active substance" and generally at least part of the dispersing agent. If sodium silicate for example has been used as dispersing agent and aluminium sulfate as the metal salt, both silica and alumina will ultimately be found on the surface of the pigment together with the "active substance". Although it does not seem possible to prove the formation of a definite compound between dispersing agent and the metal hydrous oxide, an intimate association of these two compounds takes place on the surface of the pigment. As a result the dispersing agent is irreversibly removed and thus the strongly hydrophilic nature which it conferred to the pigment for its dispersion in the aqueous medium, disappears.

The amount of metal salt to be used depends on the nature of this salt and possibly also on the nature of the dispersing agent. This amount is substantially the same as usually employed in the preparation of surface treated pigments. Appreciable results can be obtained with 0.2% of metal salt, but usually 0.5% to 3% are employed. These figures are given in weight percent of the corresponding metal oxide on the basis of dry pigment. Larger amounts may also be used such as for example 10%, but usually such large quantities do not result in further improvement of the pigmentary qualities. If a mixture of several metal salts is used these figures refer to the total amount of the corresponding oxides.

Especially in the case of titanium dioxide the formation on the pigments surface of a hydrous metal oxide and/or its products of interaction with dispersing agents such for example as alkali metal silicates or polyphosphates, results in improvements of the pigmentary properties. It is known that such improvements can be obtained under similar

224.322

conditions in the absence of an "active substance", but so prepared pigments lack organophilic properties. However, it was not obvious that this beneficial effect would still be obtained if the treatment by metal salts is done on a pigment suspension already containing an "active substance" distributed therein. It has been determined that each of the two treating agents, added in the order as indicated, produces fully its own effect without disturbing the other's action. One might have thought that the presence of the "active substance" will prevent the normal action of the added metal salt on the surface of the pigment. This is not the case, and it can be seen that in addition to its normal action the formation of the metal compound deposit contributes to a better binding of the "active substance" to the pigment particles. It is not possible to recognize on the surface of the pigment particles two distinct and successive layers, "active substance" first, then metal compounds. On the contrary it seems that these two substances are intermixed in such a manner as to reject the non polar hydrocarbon chains of the "active substance" on the surface of the treated pigment particle comprising: pigment plus "active substance" plus metal compound and dispersing agent residues. The exact manner by which these components are linked together is not well known and is not part of the invention which is only defined by the successive steps of the process in the order described and by the results obtained.

After the metal salt has been uniformly distributed through the suspension, the pigment is further processed as known in the art, that is, neutralized, washed free of soluble salts, filtered, dried and disintegrated. The drying will be done at temperatures usually applied for ordinary surface treated pigments. In some cases the temperature raise during the drying step will act favourable on the organophilic properties of the pigment. This will be the case when use is made of such "active substances" which are favourably modified by heating for instance towards a higher polymerisation - polycondensation degree as in the case of alcoylmethylol-melamines.

The process of the invention is illustrated by the following examples, but not limited to their specific descriptions:

**EXAMPLE 1.** 1.000 kg of  $\text{TiO}_2$  anatase pigment is slurried with 5.000 liters of cold water in presence of sufficient sodium silicate to raise the pH to 9 (use is made of about 15 to 25 liters of a solution containing 320 g per liter of solid and having a  $\text{SiO}_2/\text{Na}_2\text{O}$  ratio of about 3). This solution is well stirred and a solution of 5 kg sodium stearate in 250 liters of water is added thereto over a period of 15 minutes. (The sodium stearate solution may be prepared at the boiling temperature, a slight turbidity may develop on cooling but has no effect on the final results). After 10 minutes continued stirring, addition is made progressively of 1.000 kg of

---

224.322

---

hydrated aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18 \text{H}_2\text{O}$ ) dissolved in 400 liters of water. Stirring is continued for 30 minutes and then the pH is adjusted to 7 by addition of caustic soda, followed by filtering, washing and drying at  $120^\circ\text{C}$ . The finally disintegrated product is a white powder easily wetted by organic solvents such as benzene.

**EXAMPLE 2.** 1.000 kg. of a rutile  $\text{TiO}_2$  pigment (hydroclassified to a maximum particle size of 2 microns) is suspended in 5.000 liters of water in the presence of 20 liters of sodium silicate solution of identical composition as that one used in Example 1. To the thoroughly mixed slurry an aqueous solution of 5 kg sodium abietate is added. After 10 minutes continuous stirring addition is made of 130 kg of hydrated aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18 \text{H}_2\text{O}$ ) dissolved in 400 liters of water. Stirring is continued for 30 minutes and then the suspension is neutralized by means of caustic soda.

After filtering, washing, drying at  $115^\circ\text{C}$  and disintegrating a pigment is obtained, showing the same organophilic properties as that one obtained according to Example 1.

**EXAMPLE 3.** Example 2 is repeated with the difference that the aluminium sulfate solution is replaced by a mixture of 300 liters of aluminium sulfate solution having a concentration of 50 g  $\text{Al}_2\text{O}_3$  per liter and 200 liters of a zinc sulfate solution adjusted to a concentration of 25 g  $\text{ZnO}$  per liter.

The obtained pigment after drying and disintegration also shows strong organophilic properties.

**EXAMPLE 4.** 1.000 kg of  $\text{TiO}_2$  anatase pigment is dispersed in 5.000 liters of water containing about 200 liters of the sodium silicate solution referred to in Example 1 so as to obtain a final pH value of 9. When the pigment is thoroughly dispersed by stirring, addition is made to the dispersion of 20 liters of an aqueous emulsion containing 33% of silicone (in this case emulsion "sl. 35 D" marketed by the "Compagnie St. Gobain"). After 20 minutes stirring 400 liters of an aluminium sulphate solution at a concentration of 50 g  $\text{Al}_2\text{O}_3$  per liter is added progressively. Stirring is continued for 30 minutes and then the slurry is neutralized by means of caustic soda, filtered, the pigment washed, dried at  $130^\circ\text{C}$  and disintegrated. A white pigment is obtained showing strongly hydrophobic and organophilic properties. When spread on water, the pigment is not wetted but floats indefinitely on the surface. When stirred with equal quantities of water and benzene, the pigment collects entirely in the benzene layer.

**EXAMPLE 5.** The Example 4 is repeated with the same quantity of hydroclassified rutile Titanium oxide of which the particles with a diameter of over 2 microns have been eliminated. The pigment obtained

---

224,322

---

shows the same organophilic and hydrophobic characteristics as that of Example 4.

**EXAMPLE 6.** The operation described in Example 4 is repeated, but without adding the silicone emulsion. When stirred with equal quantities of water and benzene, the pigment is dispersed in the water only.

**EXAMPLE 7.** The operation described in Example 5 is repeated but without adding the silicone emulsion. In presence of water and benzene, the pigment obtained is also dispersed in water as in Example 6.

**EXAMPLE 8.** 1.000 kg of calcinated and hydroclassified  $\text{TiO}_2$  rutile are dispersed in 5.000 liters of water in the presence of 20 liters of a 300 g/l of silicate of sodium solution (having the proportion of  $\text{SiO}_2/\text{Na}_2\text{O}$  of 3:1) and the pH is adjusted to 9 by means of caustic soda. After energetic agitation to assure a good dispersion 35 liters of an aqueous emulsion of 15% polydimethylsiloxane is added over a period of 10 minutes and with thorough stirring. After 15 minutes' agitation 400 liters of a solution of sulphate of aluminium and titane assayed at 40 g/l of  $\text{Al}_2\text{O}_3$  and 25 g/l of  $\text{TiO}_2$  are added over a period of 25 minutes. When the solution of sulphates is well mixed through the mass the latter is neutralized by means of caustic soda, filtered, washed and dried at  $130^\circ\text{C}$ . The pigment obtained is strongly organophilic and hydrophobic, and, further, shows itself to have a high colouring power and exceptional resistance to farinage.

The pigmentary properties of the product obtained in the examples 1 to 8 are compared in the table 1 with the properties of rough untreated anatase (A) and untreated rutile (R) pigments used in the examples 1 to 8. These rough pigments A and R are conventionally prepared by so-called sulphate process, calcined and milled as usual.

22-1 322

Table 1.

	(A)	(R)	Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8
Brightness	12,3	11,5	12,3	11,8	11,8	13,0	12,3	13,0	12,0	12,1
Tinting Strength	135	190	135	240	230	140	250	135	230	250
Resistance to discolouration by ultra-violet light	lit.	med.	pass.	good	good	good	v.gd.	lit.	good	v.gd.
Resistance to chalking	lit.	med.	pass.	good	good	good	v.gd.	lit.	good	exc.
Resistance to marine atmos- phere	lit.	pass.	med.	good	good	v.gd.	exc.	lit.	med.	exc.
Hydrophobic properties	none	none	good	good	good	v.gd.	exc.	none	none	exc.
Resistance of paints to settling	med.	med.	good	good	good	exc.	exc.	med.	med.	exc.
Organophilic properties	lit.	lit.	good	good	good	v.gd.	exc.	lit.	lit.	exc.

---

**224.322**

---

For the same property the increasing quality is expressed by the expressions: none, little (lit.), passable (pass.), good, very good (v.gd.), excellent (exc.).

The brightness indicates a measure showing the reflectance and whiteness of the pigment, this one is the better as the figure showing the brightness is higher. The tinting strength is also the better as the representative figure is higher. The good surface treated rutile pigments have a tinting strength of 230.

The examination of Table 1 shows that the product of Example 5 is characterized by tinting strength and brightness superior to that of non-organophilic corresponding products (ex. 7) or corresponding organophilic products prepared with aliphatic fatty acid salts (ex. 2). It appears that the rutile-type Titanium dioxides treated according to our new process i.e. using silicone emulsions as "active substances" have exceptionally high tinting strength and brightness. This improvement in the tinting strength and brightness could not be expected.

This new method illustrated here by examples relating to Titanium dioxide may be successfully used for the preparation of other organophilic and hydrophobic pigments, as it is shown by the following examples.

**EXAMPLE 9.** 1.000 kg of pigmentary zinc oxide are dispersed in 8.000 liters of water by adding caustic soda as dispersing agent so as to obtain the pH 9. After good stirring a solution of 5 kg of sodium laurate in 500 liters of water is added to the dispersion. After good distribution of the sodium laurate has been reached, 300 liters of an aluminium sulphate, containing 50 g/l as  $Al_2O_3$  are added. After 15 minutes stirring the pigment is washed and, the water being filtered, it is dried at  $110^{\circ}C$  and disintegrated. The obtained pigment is easily dispersed in oils and organic solvents.

**EXAMPLE 10.** The operation of the Example 9 is repeated, but using instead of sodium laurate, 22 liters of the silicone emulsion "si 35 D" used in the Example 4. The zinc oxide which is obtained is strongly organophilic, and, when agitated in presence of equal quantities of benzene and water, it is dispersed in the organic phase only.

**EXAMPLE 11.** 1.000 kg of lithopone is suspended in 5.000 liters of water containing 4 kg of sodium pyrophosphate. After good stirring addition is made of 5 kg of hard soap (known as "marseille soap") tempered in tepid water. After 30 minutes stirring to ensure proper distribution of the soap, 400 liters of aluminium sulphate solution, containing 50 g/l expressed as  $Al_2O_3$ , are added. The pigment which is obtained, after being neutralized by sodium carbonate, washed, filtered and dried at  $110^{\circ}C$ , is characterized by its affinity for oils and organic solvents.

The claims defining the invention are as follows:

---

224.322

---

1. Process for obtaining organophilic and hydrophobic pigments, comprising dispersing the pigment in an aqueous medium in the presence of a dispersing agent, then adding to the dispersion thus obtained an active substance conferring organophilic and hydrophobic properties, then adding to the mixture obtained a soluble salt of a metal capable of forming an insoluble compound on the surface of the pigment particles and finally neutralizing, washing, filtering and drying the organophilic and hydrophobic pigment thus obtained. (28th March, 1957).
2. Process according to Claim 1, characterized in that 0.03 to 3% dispersion agent is used, and 0.1 to 5% active substance and a quantity of soluble salt of metal equivalent to 0.2 to 10% of corresponding insoluble oxide, these quantities being expressed in percentage of weight of pigment concerned. (28th March, 1957).
3. Process according to Claim 1, characterized in that a silicate of alkaline metal is used as dispersion agent. (28th March, 1957).
4. Process according to Claim 1 characterized in that a pyrophosphate of alkaline metal is used as dispersion agent. (28th March, 1957).
5. Process according to Claim 1 characterized in that a hydroxide of alkaline metal is used as dispersion agent. (28th March, 1957).
6. Process according to Claim 1 characterized in that a soluble salt of a high fatty acid is used as active substance. (28th March, 1957).
7. Process according to Claim 1 characterized in that a soluble salt of resinous acid is used as active substance. (28th March, 1957).
8. Process according to Claim 1 characterized in that an aqueous emulsion of a polyorganopolysiloxane is used as active substance, (28th March, 1957).
9. Process according to Claim 1 characterized in that a soluble salt of a metal of the group: aluminium, magnesium, zinc, titanium, zirconium, rare earth metals, tin, antimony, lead, are used in the

---

224,322

---

course of the third stage of the process. (28th March, 1957).

10. Process according to Claim 1 characterized in that soluble salts of several chosen metals in the group defined in Claim 9 are used. (28th March, 1957).

11. Process according to Claim 1 for obtaining bioxide of organophilic and hydrophobic titanium consisting of treating the anatase or calcinated and milled rutile. (28th March, 1957).

12. Process for obtaining bioxide of organophilic and hydrophobic titanium rutile having a whiteness and increased colouring power, consisting of dispersing the calcinated and ground product in water in the presence of 0.03 to 3% of silicate of sodium, by adding to the dispersion obtained 0.1 to 5% polyorganopolysiloxane in the form of an aqueous emulsion, adding to the mixture obtained a solution of salts of aluminium at 0.2 to 3% of  $Al_2O_3$  ratio to the pigment, then by neutralizing, washing, filtering and drying the bioxide of titanium thus treated. (28th March, 1957).

13. Process according to Claim 12, characterized in that in place of salts of aluminium a solution of sulphate of aluminium is used and of titanium corresponding to 0.1 to 2.5% of  $Al_2O_3$  and 0.1 to 2.5% of  $TiO_2$  proportional to the weight of pigment concerned. (28th March, 1957).

14. Process for obtaining organophilic and hydrophobic oxide of zinc according to Claim 1 characterized by using as dispersing agent caustic soda in an amount sufficient to obtain a pH value of 9 in the slurry, as active substance 0.1 to 2% by weight of polyorganopolysiloxane in the form of an aqueous emulsion and as soluble metal salt aluminium sulphate in an amount corresponding to 0.2 to 3% of  $Al_2O_3$  which percentage is based on the weight of the zinc oxide treated. (28th March, 1957).

15. Pigments treated according to the process of any one of the preceding claims. (28th March, 1957).

ARTHUR S. CAVE.  
Patent Attorney for Applicant.

\*\*\*\*\*



---

224,322

---

References:

<u>Serial No.</u>	<u>Application No.</u>	<u>Classification.</u>
165,661	23,600/53	79.2; 79.3
149,980	12,141/47	79.2; 20.2; 79.3; 47.7, 47.5
122,316	15,503/44	79.2.